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THE SEPARATION OF THE NUCLEAR ISOMERS OF TELLURIUM, MERCURY AND TIN

[This is a translation of an article by A.N. Murin, V.D. Nefedov, Ye.N. Sinotova, and O.V. Larionov in Zhurnal Neorganicheskoy Khimii, Vol VIII, No 1, Moscow, 1958, pages 181-183.]

The present communication is dedicated to the question of separating genetically-connected nuclear isomers of tellurium (Tel27), mercury (Hgl95 and Hgl97) and tin (Sn121) carried out in the Department of Radio-Chemistry in the University of Leningrad.

The method of separation of nuclear isotopes of tellurium is based on the premise that the isomer transition in tellurium is accompanied with the disintegration of the original molecule of telluric acid and the chemical stabilization of the lower isomer in the form of tellurous acid was offered by Seaborg, Livingood and Kennedy [1]. Later Williams [2] determined the yield of the lower isomer of tellurium (Tel27), which appeared to be equal to 85%. However during the separation of the lower inactive tellurous acid, the formation of the lower isomer state of tellurium without the carrier was excluded. For the separation of nuclear isomers of Tel27, we used as an original compound tellurium dimethylnitrate.

Inasmuch as the transition to the intermediate state occurs fundamentally by way of a conversion, it is possible to expect that almost every transition will be associated with a disturbance of chemical bonds of tellurium in the original compound. Therefore a considerable part of the Tel27 nuclei in the primary condition will be found in the dimethyldinitro-tellurium compound in the state of simple inorganic forms.

The method of separation of Tel21 in the primary state consisted in adsorption of these inorganic forms by means of iron hydroxide. The non-isotopic carrier -iron- was removed by extraction with isopropyl ether from 9N HCl.

From the curve of decomposition of the separated lower isomer (Fig. 1) it is apparent that only one isotope with a half-life of 9.3 hrs ($T_{\text{table}} = 9.35$ hrs) is present, and this indicates the existence in the compound of only lower isomer Tl^{127} . According to the decomposition curves of the separated compounds, the degree of contamination with the metastable Tel^{127m} was determined. In any event this degree appeared to be less than 0.1% and it could have been lowered by an additional recrystallization of the original compound. Apparently the contamination could be explained by an insufficient chemical purity of the original compound or was due to a radio effect during the storage period, inasmuch as the original compound is not capable of being absorbed by an iron hydroxide.

The determination of the output of Tl^{127} in a primary isomer state disclosed a value of 80% while it was accumulated in crystal form and 94% when accumulated in liquid solution. The latter value fully agrees with the known fact that the isomer transition into Tl^{127} is converted practically 100%. The conclusion is that with each act of isomer conversion a disintegration of the original molecule occurs, accompanied by an internal conversion.

The output during the accumulation in crystals is somewhat smaller, since in this case there is a probability of a retention at the expense of stabilization of the lower isomer in the form of the original compound. It was demonstrated that the separated Radio- Tel^{127} is present in a quadrivalent form and that only 8% is in a sixvalence-form. The cited method of separation of nuclear isomers of tellurium has a number of advantages compared to the method used by Seaborg, Livingood and Kennedy. Not being inferior to the latter from the standpoint of simplicity of the chemical method, our method offers in addition the possibility of separating the lower isomer state of the Tel^{127} nuclei without the carrier and a very high yield.

($T_{\text{exp}} \cdot \text{Hg}^{197} = 65$ hours, $T_{\text{table}} \cdot \text{Hg}^{197} = 64.5$ hrs.,

$T_{\text{exp}} \cdot \text{Hg}^{195} = 9$ hours, $T_{\text{table}} \cdot \text{Hg}^{195} = 9.5$ hours):

The following radioactive isotopes: Hg^{197} , Hg^{199} , Hg^{203} and Hg^{205} emerge as a result of the exposure of mercury to neutrons according to reactions (n, γ) and $(n, 2n)$. Since the period of time from the exposure-ending to the separation-beginning was no less than 6 days, it is possible to assume that in the original compound of the diethylmercury synthesized by us the only substances present were Hg^{203} , Hg^{197m} and Hg^{197} .

The results obtained in our laboratory during systematic investigations in the area of isotope exchange [3-6] permit us to conclude that the complete aliphatic derivatives of mercury appear in compounds in which the process of disintegration of the chemical bonds during the isomer transition will be irreversible. The separation of mercury (Hg^{197}) in the basic state was accomplished by absorbing mercury on the manganese dioxide. The separation of mercury from the carrier can be accomplished by means of methods based on the volatility of mercury and its derivatives. The fact of the separation of the nuclear isomers itself can be established by comparing the disintegration curves of the mercury compounds, obtained as a result of the disintegration of the diethylmercury and as a result of experiments with the separation of nuclear isomers, which is clear from Fig. 2. The criterion of the separation completeness would be a ratio of Hg^{197} and Hg^{203} activities. As follows from Fig. 2, the curve of the activity decline of the mercury compounds obtained as a result of the disintegrations of the diethylmercury shows the presence in the compound of only insignificant and short-lived activity belonging to nuclear isomers of Hg^{197} .

The overwhelming part of the activity stipulates Hg^{203} . Contrary to this, the activity curve of the decline of the compound obtained as a result of the nuclear isomers separation indicate the presence of only short-lived activities belonging to the basic states of Hg^{195} and Hg^{197} . The presence of Hg in a basic state in the mercury compound after the separation of the nuclear isomers was unexpected by us inasmuch as the literature points only to the possibility of obtaining Hg^{195} according to reactions $\text{Au}(d, 4n)$ and $\text{Au}(p, 3n)$. The only explanation for the presence of the basic state of Hg^{195} could be its formation from Hg^{195m} ($T=38$ hours), emerging during the exposure of mercury in the uranium reactor according to reaction: $\text{Hg}^{196}(n, 2n) \text{Hg}^{195m}$. The fact that the observed short-lived activities belong to the mercury isotopes was proved by the chemical methods of identification.

During the exposure of tin to thermal neutrons the following radioactive nuclears are formed: Sn^{113} forming during β disintegration In^{113m} , Sn^{117} , and Sn^{119m} forming during isomer transition stable isotopes Sn^{197} and Sn^{119} , Sn^{121} , Sn^{123} and Sn^{125} . For separation of Sn^{121} in a basic state from the benzine, a solution of tetraphenyl tin was used as a method of extraction. Since isomers Sn^{123} and Sn^{125} are not connected genetically, during the extraction Sn^{121} and In^{113n} will be in the liquid layer. Therefore the activity measurement was initiated after 10-12 periods of half-life of In^{113m} ($T=105$ min.).

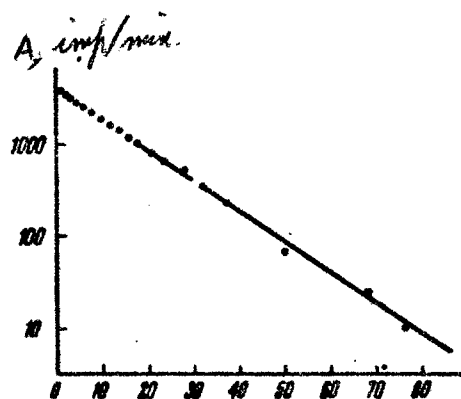


Fig. 1. Time, hours

The curve of desintegration of the lowest isomer Te^{127}

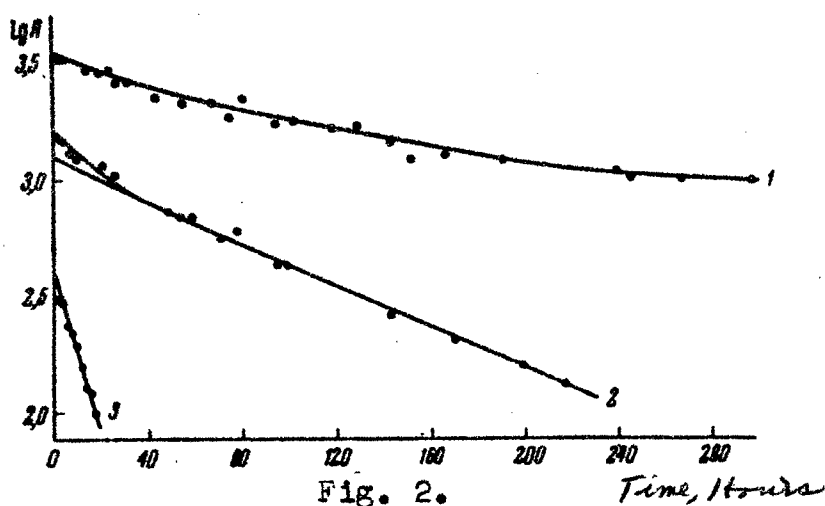


Fig. 2.

Time, hours

The curve of drop of activities of the original compound diethylmercury (1) and compound, obtained as a result of nuclear isomer separation (2). Straight line 3 obtained through splitting curve 2.

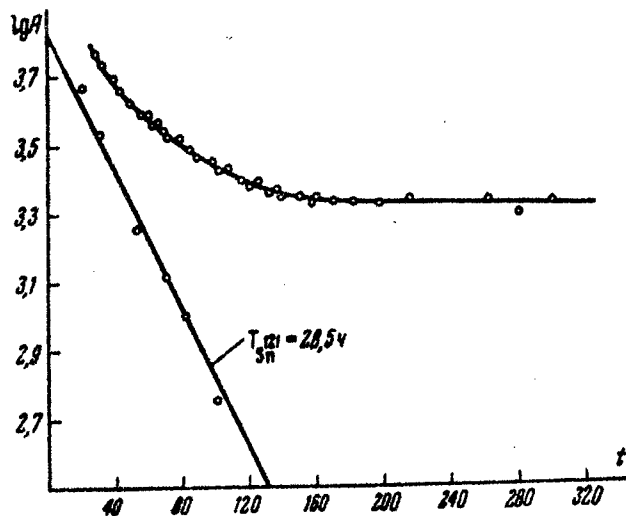


Fig. 3.
The curve of disintegration of Sn^{121}

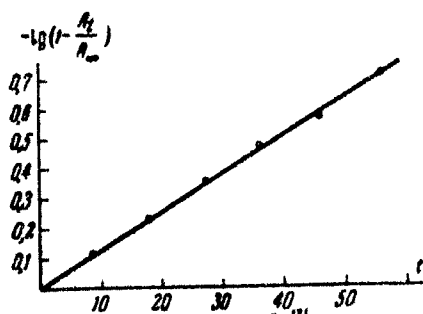


Fig. 4.
The curve of accumulation of Sn^{121} in the relation to time
accumulation of Sn^{121}

The curve of Sn^{121} disintegration is shown on Fig. 3. For the support of the genetic connection of Sn in a basic condition, with Sn^{121m} an accumulation of Sn^{121} in relation to time was studied (Fig. 4).

The cited experiments indicate that as a result of the isomer transition the chemical bonds in the molecule of a metalloorganic compound may be disturbed. A similar disturbance occurs during practically every act of the conversion of isomer transition. At the same time, in all probability the chemical form of the stabilization of the nuclear in the lower isomer state appear to be simplest inorganic compounds.

It should be noted that the method of nuclear isomer separation by means of the utilization of metalloorganic compounds may be successfully employed for a series of elements. In this sense this method can be considered as the most universal of all methods so far employed. Besides, this method allows separation of nuclei in the lowest isomer condition without the carrier.

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